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Interactive comment on "Impacts of the January 2005 solar particle event on noctilucent clouds and water at the polar summer mesopause" *by* H. Winkler et al.

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This is a very interesting paper on the impact of a solar proton event on noctilucent clouds which significantly adds to our understanding of mesospheric clouds and their environment.

My comment concerns the following statement:

'Although the temperature measurements of Lubken et al. (2009) indicate that homogeneous ice nucleation can be possible at the polar summer mesopause, it is generally believed that at typical supersaturation levels during the NLC season condensation nuclei are required for the formation of the NLC particles, (e.g., Keese, 1989; Reid, 1997).'

I disagree that it is 'generally believed' nuclei are required for the formation of NLC particles. Typical (or average) supersaturations in the upper mesopause region should not be used as a guide as to whether ice can nucleate homogeneously. Nucleation is likely to occur only at peak supersaturations which by their very nature will be transitory and therefore will only be observed rarely. In the much better studied upper troposphere supersaturations get high enough for homogeneous freezing (liquid droplet to ice) to occur, but once nucleation occurs the growth of the ice particles causes the supersaturation to reduce. Figure 3 in Kramer et al. (Atmos. Chem. Phys., 9, 3505–3522, 2009) reveals that supersaturations are rarely observed at or above that required for homogeneous freezing. If one took the mean or most likely supersaturation value you would conclude that homogeneous freezing cannot occur, which would be wrong. In the mesosphere the same is likely to be true.

Murray and Jensen (Journal of Atmospheric and Solar-Terrestrial Physics 72 (2010) 51–61) quantitatively show that transitory high supersaturations consistent with those reported by Lubken (2009) can result in homogeneous nucleation of amorphous solid water which will later transform to crystalline ice particles. It is also shown that homogeneous nucleation is possible in the presence of heterogeneous ice nuclei. The Keese (1989) and Reid (1997) references used to back up the claim that nuclei are required were written well before it was known that supersaturations in the mesopause region can reach such extreme values. In addition, those authors also made the assumption that ice nucleates directly from the vapour phase, which has been shown unambiguously not to be the case above $\sim$190 K and most likely not the case at mesospheric temperatures. Instead amorphous water (either solid or liquid depending on the temperature) will nucleate in preference because there is a smaller free energy barrier to its formation (e.g. Wolk and Strey, J. Phys. Chem. B 2001, 105, 11683-11701).

It is acknowledged that the assumption about the nucleation mechanism will not impact the modelling study directly since a constant number density is assumed at the small size (implying a constant production rate), nevertheless homogeneous nucleation should not be ruled out given present knowledge.

Other comments:

1) Equation 3 is for hexagonal ice rather than cubic ice as implied in the text. Cubic ice is known to have a greater vapour pressure than hexagonal ice with a free energy difference of 155 $\pm$ 30 J/mole (Shilling et al. GEOPHYSICAL RESEARCH LETTERS, VOL. 33, L17801, doi:10.1029/2006GL026671, 2006). The equation needed to apply this to estimate the vapour pressure under mesospheric conditions is on p 1546.

2) For which phase is the expression for surface tension (Eq 4)?

3) The conclusion that water vapour is enhance higher in the cloud layer as a result of the SPE should be mentioned explicitly in the abstract.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 1151, 2012.